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The free energy of quantum spin systems: Functional integral representation

Peter Wölfle,¹ Natalia B. Perkins,² and Yuriy Sizyuk²

¹*Institute for Condensed Matter Theory and Institute for Nanotechnology,
Karlsruhe Institute of Technology, D-76128 Karlsruhe, Germany*

²*School of Physics and Astronomy, University of Minnesota, Minneapolis, MN 55116, USA*

In this work, we propose a method for calculating the free energy of anisotropic quantum spin systems. We use the Hubbard-Stratonovich transformation to express the partition function of a generic bilinear super-exchange Hamiltonian in terms of a functional integral over classical time-dependent fields. In the general case the result is presented as a product of traces over single spins subject to a time-dependent field. The traces may be evaluated in closed form in the case of Ising-type spin systems. In the General case we derive a compact expression for the contribution of Gaussian spin fluctuations to the free energy. We show how anisotropic spin interactions lead to anisotropies in the free energy, giving rise to pinning of the spontaneous magnetization along preferred directions

I. INTRODUCTION

Recent research activities on $4d$ and $5d$ transition metal oxides have shown that the interplay of the strong spin-orbit coupling (SOC), crystal field interactions, and electron correlations may lead to strongly anisotropic, bond-dependent exchange interactions between localized magnetic moments.^{1–15} These anisotropic interactions have the generic form $J_{j,j'}^{\alpha\alpha'} S_j^\alpha S_{j'}^{\alpha'}$, in which S_j^α denotes the α -th component of the pseudospin operator on site j , describing the magnetic degree of freedom, \mathcal{J}_{eff} . The most notable examples appear in iridium and ruthenium systems,¹⁶ in which magnetic degrees of freedom on Ir^{4+} or Ru^{3+} ions arise from a low-lying multiplet (usually a doublet) of total angular momentum $\mathcal{J}_{\text{eff}} = \mathcal{L}_{\text{eff}} + \mathcal{S} = 1/2$, in which spin and orbital angular momenta are intertwined due to the strong SOC.

In systems with strong SOC, the exchange interactions $J_{j,j'}^{\alpha\alpha'}$ are generally not $\text{SU}(2)$ invariant and are lattice-specific, because magnetic moments contain both spin and orbital angular momentum components. The presence of anisotropic bond-directional spin interactions in SOC models provides the foundation for the realization of a plethora of novel quantum ground states, such as the Kitaev spin liquid,¹⁷ and a rich variety of magnetically ordered states^{18–21} including single- and multiple- Q spirals.^{22–24} While the theoretical analysis of the structure of magnetic ground states of the SOC magnets has recently received considerable attention, only few investigations have addressed the problem of how the direction of the order parameter is selected.^{9–13}

There are two possible scenarios for the selection of the direction of the order parameter. In the most generic anisotropic biquadratic models, it might be selected already at the mean field level by off-diagonal pseudospin interactions even if these interactions are subdominant with respect to diagonal couplings.^{11,25} In the exchange models with only diagonal interactions, i.e. in the compass-like models, the magnetic orders with different directions of the order parameters are degenerate on the mean field level. However, this classical degeneracy is

accidental and is therefore lifted by the order by disorder mechanism which selects a discrete set of states, each with a particular direction of the order parameter.

The thermal²⁶ and quantum order-by-disorder²⁷ mechanisms selecting a particular spin ordering pattern from a classically degenerate manifold of states have been proposed to be at play in a number of condensed matter systems.^{12,28–33} However, in most of the cases the quantum fluctuations are considered only at zero temperature, while at finite temperatures only classical spin fluctuations are considered.

The purpose of this paper is to present a general approach to compute the free energy of quantum spin systems with anisotropic interactions and study how spin fluctuations explicitly break the degeneracy at the mean-field level and select a particular direction of the order parameter from the manifold of classically degenerate states at finite temperatures. The formal procedure which we will be using here is based on the derivation of the fluctuation-induced part of the free energy on top of the mean field contribution, which then allows to determine the orientations of the vector order parameter corresponding to the free energy minima.

In general, the calculation of the free energy of a quantum spin system is complicated by the fact that the spin operators are non-canonical, which limits the usefulness of renormalized perturbation theory, the usual tool in dealing with quantum many-body systems.^{37,38} Here we explore a different approach: decoupling of the bilinear interaction operators by way of Hubbard-Stratonovich (HS) transformations.^{39,40} Our approach differs from a previous derivation^{34–36} of a path integral representation of interacting quantum spin systems in terms of Hubbard-Stratonovich fields in that it applies to anisotropic spin systems and is not restricted to ultralow temperatures but allows to calculate the contribution of Gaussian fluctuations to the free energy at all temperatures in the ordered phase. This requires introducing space and time-dependent HS-fields, which are integrated over. The resulting quantum trace over an exponential involving spin operators (a kind of Zeeman interaction of spins with the HS-induced "magnetic field"),

may be done explicitly. The result is a representation of the partition function in terms of an infinite power series in the interaction. The series may be summed up explicitly in the case of an Ising-type interaction or in the case of classical spins.^{25,41} In the general case of the bilinear interaction of quantum spins, only the Gaussian fluctuation contribution to the free energy of an ordered state may be derived in a compact form. Higher order corrections are accessible, but involve increasingly complex expressions.

This paper is organized as follows: In Section II, the representation of the partition function is introduced. In Section III we present the evaluation of the free energy in the approximation of Gaussian fluctuations about the mean field order parameter. We apply the derived formalism to the quantum Heisenberg-compass spin model in section IV. Finally, we draw conclusions in Section V.

II. REPRESENTATION OF THE PARTITION FUNCTION

We consider a generic anisotropic quantum spin model on a lattice defined by the Hamiltonian

$$\mathcal{H} = \frac{1}{2} \sum_{j,j'} \sum_{\alpha\alpha'} J_{j,j'}^{\alpha\alpha'} S_j^\alpha S_{j'}^{\alpha'}, \quad (1)$$

where j, j' are combined indices, $j = (i, \mu)$, denoting the position of the site by the position of the unit cell i and sublattice index μ inside the unit cell. $\alpha, \alpha' = x, y, z$ label the three components of the spin. In principle the \mathbf{S}_j may be general spin operators, but we will henceforth assume $S = 1/2$, which is the most important case. For the models with compasslike anisotropic and Heisenberg isotropic interactions of spins, the interaction is diagonal in spin space, $J_{j,j'}^{\alpha\alpha'} \propto \delta_{\alpha\alpha'}$. However, since our consideration is also valid for the case when $\alpha \neq \alpha'$, in the following, we will keep both indices.

The partition function of the system is given by the trace over many-body spin space of the Boltzmann operator

$$Z = \text{Tr} \left[\exp \left[-\beta \frac{1}{2} \sum_{j,j'} \sum_{\alpha\alpha'} J_{j,j'}^{\alpha\alpha'} S_j^\alpha S_{j'}^{\alpha'} \right] \right], \quad (2)$$

where $\beta = 1/k_B T$ is the inverse temperature (we will use units with Boltzmann's constant $k_B = 1$ and also Planck's constant $\hbar = 1$).

It is useful to represent the Hamiltonian in the basis of the normalized eigenfunctions, $\chi_{\mathbf{q},\nu;j,\alpha}$, of the spin exchange matrix $J_{j,j'}^{\alpha\alpha'}$. Here we denote the lattice vectors as $\mathbf{R}_j = \mathbf{R}_i + \tau_\mu$, where $i = 1, \dots, N$ specifies the unit cell and τ_μ , $\mu = 1, \dots, N_s$ denotes the lattice vectors inside a unit cell. The eigenfunctions $\chi_{\mathbf{q},\nu}$ and eigenvalues $\kappa_{\mathbf{q},\nu}$ are defined as

$$\sum_{j',\alpha'} J_{j,j'}^{\alpha\alpha'} \chi_{\mathbf{q},\nu;j',\alpha'} = \kappa_{\mathbf{q},\nu} \chi_{\mathbf{q},\nu;j,\alpha} \quad (3)$$

For spins on a periodic lattice these eigenstates are labeled by a wavevector \mathbf{q} from the first Brillouin zone (BZ) and index ν , whose dimensionality depends on the number N_s of non-equivalent atoms in the magnetic unit cell, $\nu = 1, \dots, 3N_s$. Explicitly we have $\chi_{\mathbf{q},\nu;j,\alpha} = N^{-1/2} \exp(i\mathbf{q} \cdot \mathbf{R}_i) u_{\mathbf{q}\nu}^{\mu\alpha}$, where the $u_{\mathbf{q}\nu}^{\mu\alpha}$ are the $3N_s$ components (labeled $\mu\alpha$) of the normalized eigenvector labeled by $\mathbf{q}\nu$ and N is the number of lattice cells. Defining collective spin operators $\tilde{S}_{\mathbf{q},\nu}$ we can express the Hamiltonian as

$$\mathcal{H} = \sum_{\mathbf{q},\nu} \kappa_{\mathbf{q},\nu} \tilde{S}_{\mathbf{q},\nu}^* \tilde{S}_{\mathbf{q},\nu}, \quad (4)$$

$$\tilde{S}_{\mathbf{q},\nu} = \sum_{j,\alpha} \chi_{\mathbf{q},\nu;j,\alpha}^* S_j^\alpha,$$

where $\tilde{S}_{\mathbf{q}}$ is a $3N_s$ -component spin vector, $\tilde{S}_{\mathbf{q},\nu}^* = \tilde{S}_{-\mathbf{q},\nu}$, and $\kappa_{\mathbf{q},\nu}$ is the diagonal interaction matrix in momentum space, which is obtained by Fourier transformation of the interaction matrix $J_{j,j'}^{\alpha\alpha'}$, and with the help of a unitary transformation in the $3N_s$ dimensional space of spin components α and unit cell positions μ . From the symmetry of the exchange interaction, $J_{j,j'}^{\alpha\alpha'} = J_{j',j}^{\alpha'\alpha}$ it follows that $\kappa_{\mathbf{q},\nu} = \kappa_{-\mathbf{q},\nu}$. For simplicity, in the following we will omit the tilde sign and put $S_{\mathbf{q},\nu} \equiv \tilde{S}_{\mathbf{q},\nu}$.

In the eigenmode representation, the partition function of the system is given by

$$Z = \text{Tr} \left[\exp \left[-\beta \sum_{\mathbf{q},\nu} \kappa_{\mathbf{q},\nu} S_{\mathbf{q},\nu}^* S_{\mathbf{q},\nu} \right] \right], \quad (5)$$

A. Hubbard-Stratonovich transformation

Our next step is to apply the Hubbard-Stratonovich transformation to each normal component separately. This transformation is based on the mathematical identity (defining $x = \text{Re}\{S_{\mathbf{q},\nu}\}$, $y = \text{Im}\{S_{\mathbf{q},\nu}\}$, and $a = \beta\kappa_{\mathbf{q},\nu}$)

$$\exp[-a(x^2 + y^2)] = \frac{1}{\pi|a|} \int du dv \quad (6)$$

$$\begin{cases} \exp \left[-\frac{u^2 + v^2}{|a|} + 2(xu + yv) \right], & a < 0, \\ \exp \left[-\frac{u^2 + v^2}{|a|} + 2i(-xu + yv) \right], & a > 0 \end{cases},$$

In the second equality we made use of the freedom to choose the imaginary prefactor to be $+i$ or $-i$. Here the auxiliary variables u, v are components of the Hubbard-Stratonovich field $\varphi_{\mathbf{q},\nu}$, which we choose to be $u = \text{Re} \varphi_{\mathbf{q},\nu}$ and $v = \text{Im} \varphi_{\mathbf{q},\nu}$ in the first equation (valid if $a < 0$) and $v = \text{Re} \varphi_{\mathbf{q},\nu}$ and $u = \text{Im} \varphi_{\mathbf{q},\nu}$ in the second equation (valid if $a > 0$). We may combine both cases by defining a function $s(\kappa_{\mathbf{q},\nu}) = 1$ if $\kappa_{\mathbf{q},\nu} < 0$ and $s(\kappa_{\mathbf{q},\nu}) = -i$ if $\kappa_{\mathbf{q},\nu} > 0$. Then in the first equation $(xu + yv) = \text{Re}\{s(\kappa_{\mathbf{q},\nu}) S_{\mathbf{q},\nu}^* \varphi_{\mathbf{q},\nu}\}$, whereas in the second equation $i(-xu + yv) = \text{Im}\{S_{\mathbf{q},\nu} \varphi_{\mathbf{q},\nu}^*\}$, which may also

be expressed as $\text{Re}\{s(\kappa_{\mathbf{q},\nu})S_{\mathbf{q},\nu}\varphi_{\mathbf{q},\nu}^*\}$. In both cases, whether $s(\kappa_{\mathbf{q},\nu})$ is real or imaginary valued, the term in the exponent linear in $S_{\mathbf{q},\nu}$ is real valued. In Eq.(6), $x+iy$ may be a number or an operator.

The application of the Hubbard-Stratonovich transformation to (5) requires the normal components of the spin operators to commute with each other, which is true for classical spins. Then using the Hubbard-Stratonovich transformation one may express the Boltzmann weight operator of each normal mode in terms of classical fields $\varphi_{\mathbf{q},\nu}$ and represent the interaction operator as a Zeeman energy operator of spins in a spatially varying magnetic field⁴¹. Note that in⁴¹ we used a slightly different convention for the definition of variables u, v above.

In order to make use of the Hubbard-Stratonovich transformation for the computation of the partition function (5) of a quantum spin system, for which the $S_{\mathbf{q},\nu}$ do not commute, we need to represent the Boltzmann operator as an evolution operator in imaginary time and apply the Suzuki-Trotter discretization⁴², allowing to write down the partition function in terms of products over time slices.³⁷ Explicitly, we have

$$\begin{aligned} Z &= \text{Tr} [\exp(-\beta\mathcal{H})] \\ &= \text{Tr} \left[T_\tau \exp\left[-\epsilon \sum_{n=1}^M \mathcal{H}(\tau_n)\right] \right], \end{aligned} \quad (7)$$

where T_τ is the imaginary time ordering operator, and we sliced the imaginary time interval $[0, \beta]$ into $M = \beta/\epsilon$ infinitesimal intervals each of length ϵ , and $\tau_n = n\epsilon$, $n = 1, 2, \dots, M$. Since ϵ is small, and will be taken to zero at

the end, we may now expand each exponent in Eq. (7) as

$$\exp[-\epsilon\mathcal{H}(\tau_n)] = 1 - \epsilon\mathcal{H}(\tau_n) + O(\epsilon^2), \quad (8)$$

and approximate

$$T_\tau \exp\left[-\epsilon \sum_{n=1}^M \mathcal{H}(\tau_n)\right] = T_\tau \prod_{n=1}^M \exp[-\epsilon\mathcal{H}(\tau_n)], \quad (9)$$

where

$$\begin{aligned} \exp[-\epsilon\mathcal{H}(\tau_n)] &= \exp\left[-\epsilon \sum_{\mathbf{q},\nu} \kappa_{\mathbf{q},\nu} S_{\mathbf{q},\nu}^*(\tau_n) S_{\mathbf{q},\nu}(\tau_n)\right] \\ &= \prod_{\mathbf{q},\nu} \exp[-\epsilon \kappa_{\mathbf{q},\nu} S_{\mathbf{q},\nu}^*(\tau_n) S_{\mathbf{q},\nu}(\tau_n)] + O(\epsilon^2). \end{aligned} \quad (10)$$

Now, at each instant τ_n we may perform the Hubbard-Stratonovich transformation separately and express the Boltzmann weight operator of each normal mode in terms of normal field amplitudes $\varphi_{\mathbf{q},\nu}(\tau_n)$ as

$$\begin{aligned} \exp[-\epsilon \kappa_{\mathbf{q},\nu} S_{\mathbf{q},\nu}^*(\tau_n) S_{\mathbf{q},\nu}(\tau_n)] &= \\ C_{\mathbf{q},\nu}^{-1} \int d\varphi_{\mathbf{q},\nu}^* d\varphi_{\mathbf{q},\nu} \exp\left[-\epsilon \left\{ |\kappa_{\mathbf{q},\nu}|^{-1} \varphi_{\mathbf{q},\nu}^*(\tau_n) \varphi_{\mathbf{q},\nu}(\tau_n) \right. \right. \\ &\quad \left. \left. - 2 \text{Re}\{s(\kappa_{\mathbf{q},\nu}) S_{\mathbf{q},\nu}^*(\tau_n) \varphi_{\mathbf{q},\nu}(\tau_n)\} \right\} \right]. \end{aligned} \quad (11)$$

The partition function (7) may therefore be expressed as

$$\begin{aligned} Z &= \frac{1}{C} \text{Tr} \left[T_\tau \int [d\varphi] \prod_{n=1}^M \exp \left[-\epsilon \left\{ \sum_{\mathbf{q},\nu} (|\kappa_{\mathbf{q},\nu}|^{-1} \varphi_{\mathbf{q},\nu}^*(\tau_n) \varphi_{\mathbf{q},\nu}(\tau_n) - 2 \text{Re}\{s(\kappa_{\mathbf{q},\nu}) S_{\mathbf{q},\nu}^*(\tau_n) \varphi_{\mathbf{q},\nu}(\tau_n)\} \right\} \right] \right] \\ &= \frac{1}{C} \int [d\varphi] \prod_{n=1}^M \exp \left[-\epsilon \sum_{\mathbf{q},\nu} |\kappa_{\mathbf{q},\nu}|^{-1} \varphi_{\mathbf{q},\nu}^*(\tau_n) \varphi_{\mathbf{q},\nu}(\tau_n) \right] \text{Tr} \left[T_\tau \prod_{j,\alpha} \exp[-\epsilon B_{j,\text{eff}}^\alpha(\tau_n) S_j^\alpha(\tau_n)] \right], \end{aligned} \quad (12)$$

where the spatially and "temporally" varying local magnetic field $B_{j,\text{eff}}^\alpha(\tau_n)$ is defined by

$$\begin{aligned} B_{j,\text{eff}}^\alpha(\tau_n) &= -2\varphi_j^\alpha(\tau_n) \\ \varphi_j^\alpha(\tau_n) &= \sum_{\mathbf{q},\nu=1}^{3N_s} \text{Re}\{s(\kappa_{\mathbf{q},\nu}) \chi_{\mathbf{q},\nu;j,\alpha}^* \varphi_{\mathbf{q},\nu}(\tau_n)\}. \end{aligned} \quad (13)$$

In Eq. (12), we also defined the integration volume element as

$$[d\varphi] = \prod_{\mathbf{q},\nu,n} d\varphi_{\mathbf{q},\nu}^*(\tau_n) d\varphi_{\mathbf{q},\nu}(\tau_n)$$

and the normalization factor C as

$$C = \int [d\varphi] \prod_{n=1}^M \exp \left[-\epsilon \sum_{\mathbf{q},\nu} |\kappa_{\mathbf{q},\nu}|^{-1} \varphi_{\mathbf{q},\nu}^*(\tau_n) \varphi_{\mathbf{q},\nu}(\tau_n) \right]$$

B. Trace over quantum spin states

We may now perform the trace over the quantum spin states. As spin operators on different sites commute, the trace may be split up into a product of traces Tr_j over

single spin Hilbert space

$$Z = \frac{1}{C'} \int [d\varphi] \exp \left[- \int_0^\beta d\tau \sum_{\mathbf{q}, \nu} |\kappa_{\mathbf{q}, \nu}|^{-1} \varphi_{\mathbf{q}, \nu}^*(\tau) \varphi_{\mathbf{q}, \nu}(\tau) \right] \prod_j \Theta_j, \quad (14)$$

where we defined

$$\Theta_j = \frac{1}{2} \text{Tr}_j \left\{ T_\tau \prod_n \exp \left[2\epsilon \sum_{\alpha=x,y,z} \varphi_j^\alpha(\tau_n) S_j^\alpha \right] \right\}. \quad (15)$$

The factors $\frac{1}{2}$ in front of the trace are compensated by corresponding factors in the normalization, leading to $C' = C/2^{N_t}$, where N_t is the total number of spins.

We note that the factors under the trace, $\exp[2\epsilon \varphi_j(\tau_n) \cdot \mathbf{S}_j]$, may be cyclically permuted. This suggests that the terms at $n = 1$ and at $n = M$ should be equal, in order to avoid an unphysical discontinuity when passing from $n = 1$ to $n = M$. In other words, we assume periodic boundary conditions, $\varphi_{\mathbf{q}, \nu}(\tau_n) = \varphi_{\mathbf{q}, \nu}(\tau_n + \beta)$. This implies that the Fourier frequencies ω_n of $\varphi_{\mathbf{q}, \nu}(\tau)$ are bosonic Matsubara frequencies.

The spin trace may be performed by inserting suitable representations of the unit operator in single spin space at each time step

$$\hat{1} = \sum_{s=\pm 1} |s; \mathbf{e}_n\rangle \langle s; \mathbf{e}_n|, \quad (16)$$

where $\mathbf{e}_n \equiv \mathbf{e}(\tau_n) = \varphi_j(\tau_n)/|\varphi_j(\tau_n)|$ denotes the spin quantization axis at time τ_n and $|s; \mathbf{e}_n\rangle \equiv |s_n\rangle$, $s = \pm 1$ are corresponding two-component spin eigenvectors, obeying $(\mathbf{S} \cdot \mathbf{e}_n)|s_n\rangle = \frac{s}{2}|s_n\rangle$. The assumed periodic boundary condition implies that $\mathbf{e}_N = \mathbf{e}_1$ such that $\mathbf{e}(\beta) = \mathbf{e}(0)$ and $\mathbf{e}(\tau)$ is a periodic function of period β .

Now, inserting the unit operator expressed in the basis of eigenstates of each factor $e^{\epsilon \varphi_n \cdot \boldsymbol{\sigma}}$ to the left and right of that factor in the Eq. 15, we may express Θ_j as

$$\begin{aligned} \Theta &= \frac{1}{2} \text{Tr} \left\{ e^{\epsilon \varphi_N \cdot \boldsymbol{\sigma}} \dots e^{\epsilon \varphi_2 \cdot \boldsymbol{\sigma}} e^{\epsilon \varphi_1 \cdot \boldsymbol{\sigma}} \right\} \\ &= \frac{1}{2} \sum_{s_1, s'_1, s_2, \dots} \langle s_1 | e^{\epsilon \varphi_N \cdot \boldsymbol{\sigma}} | s'_N \rangle \langle s'_N | \dots \\ &\quad \dots | s'_2 \rangle \langle s'_2 | e^{\epsilon \varphi_2 \cdot \boldsymbol{\sigma}} | s_2 \rangle \langle s_2 | s'_1 \rangle \langle s'_1 | e^{\epsilon \varphi_1 \cdot \boldsymbol{\sigma}} | s_1 \rangle, \end{aligned} \quad (17)$$

where for brevity we suppressed the site index j and defined $\varphi_n = \varphi(\tau_n)$, where $\varphi = (\varphi^x, \varphi^y, \varphi^z)$ is the vector in spin space.

Next, we need to compute the matrix elements $\langle s'_n | e^{\epsilon \varphi_1 \cdot \boldsymbol{\sigma}} | s_n \rangle$ and the inner products $\langle s_{n+1} | s'_n \rangle$. The former are diagonal by construction:

$$\langle s'_n | e^{\epsilon \varphi_n \cdot \boldsymbol{\sigma}} | s_n \rangle = \delta_{s_n s'_n} \exp[\epsilon \varphi_n^z s_n], \quad (18)$$

where $\varphi_n = |\varphi(\tau_n)| = \sqrt{(\varphi^x)^2 + (\varphi^y)^2 + (\varphi^z)^2}$. The inner product is given by

$$\begin{aligned} \langle s_{n+1} | s'_n \rangle &= \langle s_{n+1}, \mathbf{e}_{n+1} | s'_n, \mathbf{e}_n \rangle \\ &= \langle s_{n+1} | \exp[-i(\mathbf{e}_{n+1} \times \mathbf{e}_n) \cdot \boldsymbol{\sigma}] | s'_n, \mathbf{e}_{n+1} \rangle \\ &= \delta_{s_{n+1} s'_n} - i(\mathbf{e}_{n+1} \times \mathbf{e}_n) \cdot \boldsymbol{\sigma}_{s_{n+1} s'_n} + O(\epsilon^2). \end{aligned} \quad (19)$$

Next we define the infinitesimal angle of rotation of the quantization axis \mathbf{e}_{n+1} into \mathbf{e}_n as $\epsilon \boldsymbol{\Omega}_{n+1, n} = (\mathbf{e}_n \times \mathbf{e}_{n+1})$ and express Θ as

$$\begin{aligned} \Theta &= \frac{1}{2} \text{Tr} \{ e^{\epsilon \varphi_N \cdot \boldsymbol{\sigma}} e^{i\epsilon \boldsymbol{\Omega}_{1, N-1} \cdot \boldsymbol{\sigma}} e^{\epsilon \varphi_{N-1} \cdot \boldsymbol{\sigma}} \dots \\ &\quad \dots e^{i\epsilon \boldsymbol{\Omega}_{3, 2} \cdot \boldsymbol{\sigma}} e^{\epsilon \varphi_2 \cdot \boldsymbol{\sigma}} e^{i\epsilon \boldsymbol{\Omega}_{2, 1} \cdot \boldsymbol{\sigma}} e^{\epsilon \varphi_1 \cdot \boldsymbol{\sigma}} \}, \end{aligned} \quad (20)$$

where we denote the quantization axis at time $\tau = 0$ as $\mathbf{e}_1 = \mathbf{e}(0) = \hat{z}$. In the continuum approximation, we have

$$\mathbf{e}_{n+1} = \mathbf{e}(\tau_{n+1}) = \mathbf{e}(\tau_n) + \epsilon \dot{\mathbf{e}}(\tau_n) + O(\epsilon^2), \quad (21)$$

where $\dot{\mathbf{e}}(\tau) = \partial \mathbf{e} / \partial \tau$.

The factors in the product on the r.h.s. of the Eq.(20) commute under the time ordering operator, so we may now express Θ as

$$\Theta = \frac{1}{2} \text{Tr} \left\{ T_\tau \exp \left[\int_0^\beta d\tau \{ i\boldsymbol{\Omega}(\tau) + \varphi(\tau) \mathbf{e}(0) \} \cdot \boldsymbol{\sigma} \right] \right\}, \quad (22)$$

where $\boldsymbol{\Omega}(\tau_n) = \boldsymbol{\Omega}_{n+1, n} = \mathbf{e}(\tau_n) \times \dot{\mathbf{e}}(\tau_n)$. The term $i\boldsymbol{\Omega}(\tau)$ may be identified with the well-known Berry phase term.

Although a general evaluation of this expression involves only a trace over a single spin, it appears to be difficult. We therefore consider the following approximations. The field $\boldsymbol{\Omega}$ is by definition a fluctuation field, i.e. it vanishes on the mean field level. It therefore makes sense to expand Θ in terms of $\boldsymbol{\Omega}$. In zeroth order we have

$$\begin{aligned} \Theta^{(0)} &= \frac{1}{2} \text{Tr} \{ e^{\epsilon \varphi_N \cdot \boldsymbol{\sigma}} e^{\epsilon \varphi_{N-1} \cdot \boldsymbol{\sigma}} \dots e^{\epsilon \varphi_2 \cdot \boldsymbol{\sigma}} e^{\epsilon \varphi_1 \cdot \boldsymbol{\sigma}} \} \\ &= \frac{1}{2} \text{Tr} \{ e^{\int d\tau \varphi(\tau) \cdot \boldsymbol{\sigma}} \} = \cosh \beta \varphi_0, \end{aligned} \quad (23)$$

where the time average φ_0 is defined as

$$\varphi = \beta^{-1} \int_0^\beta d\tau \varphi(\tau). \quad (24)$$

We note for later that $\Theta^{(0)}$ contains contributions from both longitudinal and transverse fluctuations about the mean field configuration.

In first order in $\boldsymbol{\Omega}$ we find

$$\begin{aligned} \Theta^{(1)} &= \frac{1}{2} i\epsilon \sum_{n_0=1}^{N-1} \text{Tr} \{ e^{\epsilon \sigma^z \sum_{n_2=n_0+1}^N \varphi_{n_2}} [\boldsymbol{\Omega}_{n_0+1, n_0} \cdot \boldsymbol{\sigma}] \\ &\quad \times e^{\epsilon \sigma^z \sum_{n_1=1}^{n_0} \varphi_{n_1}} \}. \end{aligned} \quad (25)$$

Higher order contributions in Ω may be derived but they lead to increasingly complicated expressions. In the continuum approximation we may express $\Theta^{(1)}$ as

$$\Theta^{(1)} = \frac{i}{2} \int d\tau_0 \text{Tr} \{ e^{\sigma^z \Phi(\beta, \tau_0)} [\mathbf{\Omega}(\tau_0) \cdot \boldsymbol{\sigma}] e^{\sigma^z \Phi(\tau_0, 0)} \}, \quad (26)$$

where $\Phi(\beta, \tau_0) = \int_{\tau_0}^{\beta} d\tau \varphi(\tau)$ has been defined. We now recall that $\sigma^z = \mathbf{e}(0) \cdot \boldsymbol{\sigma}$ and use $e^{\sigma^z \Phi} = \cosh \Phi + \sigma^z \sinh \Phi$ to get

$$\begin{aligned} \Theta^{(1)} = i \int d\tau_0 [& \cosh \Phi(\beta, \tau_0) \sinh \Phi(\tau_0, 0) \\ & + \sinh \Phi(\beta, \tau_0) \cosh \Phi(\tau_0, 0)] [\mathbf{\Omega}(\tau_0) \cdot \mathbf{e}(0)], \end{aligned} \quad (27)$$

where we also used $\text{Tr} \{ \boldsymbol{\sigma} \} = 0$, $\text{Tr} \{ \sigma^z \sigma^\lambda \} = 2\delta_{\lambda, z} = \text{Tr} \{ \sigma^\lambda \sigma^z \}$ and $\text{Tr} \{ \sigma^z \sigma^\lambda \sigma^z \} = 0$. Using another identity, $\sinh x \cosh y + \cosh x \sinh y = \sinh(x + y)$, and the periodic boundary condition relations $\Phi(\beta, \tau_0) + \Phi(\tau_0, 0) = \Phi(\beta, 0) = \beta\varphi_0$ we get

$$\Theta^{(1)} = i[\mathbf{\Omega}_0 \cdot \mathbf{e}(0)] \sinh(\beta\varphi_0), \quad (28)$$

where $\mathbf{\Omega}_0 \equiv \int d\tau_0 \mathbf{\Omega}(\tau_0)$.

The partition function Eq. (14), which includes the first order correction in $\mathbf{\Omega}_0$, is then given by

$$Z = \frac{1}{C'} \int [d\varphi] \exp[-\beta(\mathcal{S}_\kappa + \mathcal{S}_{loc} + \mathcal{S}_0)], \quad (29)$$

where the interaction part of the action is given by

$$\mathcal{S}_\kappa = \beta^{-1} \int_0^\beta d\tau \sum_{\mathbf{q}, \nu} |\kappa_{\mathbf{q}, \nu}|^{-1} \varphi_{\mathbf{q}, \nu}^*(\tau) \varphi_{\mathbf{q}, \nu}(\tau) \quad (30)$$

and the local part of the action $\mathcal{S}_{loc} = \mathcal{S}_{loc}^{stat} + \mathcal{S}_{loc}^{dyn}$ has both static and dynamic contributions:

$$\mathcal{S}_{loc}^{stat} = -\beta^{-1} \sum_j \ln \cosh(\beta\varphi_{j,0}) \quad (31)$$

$$\mathcal{S}_{loc}^{dyn} = -i\beta^{-1} \sum_j \tanh(\beta\varphi_{j,0}) [\mathbf{\Omega}_{j,0} \cdot \mathbf{e}_j(0)], \quad (32)$$

and $\mathcal{S}_0 = \beta^{-1} \ln C'$.

Our results in Eqs.(29-32) agree with those of Ref.³⁴ except for an additional term involving the product of two time derivatives of the transverse field components at equal times. Such a term arises from expansion of Eq.(22) in second order in Ω , if the time arguments are kept equal. The fluctuations we will be interested in (e.g. spin waves) are long-range correlated in time such that it does not make sense to single out only the equal time products of Ω . We also note that Angelucci and Jug³⁴ did not pay attention to the fact that the Hubbard-Stratonovich transformation changes its character if the eigenvalues $\kappa_{\mathbf{q}, \nu}$ of the interaction kernel change sign.

III. MEAN FIELD SOLUTION

In this section, we consider the simplest case and compute the mean field free energy for the range of parameters of a model (1), for which the mean-field solution is a collinear magnetic state. In this case we can write $\varphi_{j,\mu}(\tau) = \varphi_{\text{MF}} \hat{\mathbf{m}}$, where $\hat{\mathbf{m}}$ is a normalized 3-component vector pointing in the direction of the spontaneous magnetization, which is the same for all sublattice sites μ . The trace in spin space is obtained as

$$\Theta_{j,\mu}^{\text{MF}} = \frac{1}{2} \text{Tr}_j \{ e^{-\beta\varphi_{\text{MF}} [\boldsymbol{\sigma} \cdot \mathbf{m}]} \} = \cosh(\beta\varphi_{\text{MF}}),$$

where the mean field expression for the fields $\varphi_{\mathbf{q}, \nu}(\tau)$ is given by $\varphi_{\mathbf{q}, \nu}^{MF} = (NN_s)^{1/2} \delta_{\mathbf{q}, 0} \varphi_{\text{MF}} m_{\mathbf{q}, \nu}$, where N is the total number of unit cells and N_s is three times the number of the sublattices in the unit cell. The normalized unit vector $m_{\mathbf{q}, \nu} = N_s^{-1/2} \sum_{\mu, \alpha} m^\alpha u_{\mathbf{q}, \nu}^{\mu\alpha}$ is expressed in terms of the eigenvectors $u_{\mathbf{q}, \nu}^{\mu\alpha}$. The mean-field partition function (14) can be easily evaluated and equals

$$Z^{\text{MF}} = \frac{1}{C'} \exp \left[-NN_s \left\{ \beta |\kappa_0|^{-1} \varphi_{\text{MF}}^2 - \ln(\cosh(\beta\varphi_{\text{MF}})) \right\} \right], \quad (33)$$

provided $\kappa_{\mathbf{q}=0, \nu} = \kappa_0$ is independent of ν . Here φ_{MF} is the value minimizing the free energy $F^{\text{MF}} = -\beta^{-1} \ln Z^{\text{MF}}$ and is given by the solution of the transcendental equation

$$2|\kappa_0|^{-1} \varphi_{\text{MF}} = \tanh(\beta\varphi_{\text{MF}}). \quad (34)$$

The full partition function is given by

$$Z = \frac{Z^{\text{MF}}}{C'} \int [d\delta\varphi] \exp[-\beta\delta\mathcal{S}],$$

where $\delta\mathcal{S} = \delta\mathcal{S}^{stat} + \delta\mathcal{S}^{dyn}$ is the fluctuational part of the action.

Despite the anisotropic form of the interactions in the Hamiltonian(1), the mean-field solution is highly degenerate with respect to the orientation of the spontaneous magnetization vector $\hat{\mathbf{m}}$. It is therefore of interest to calculate the corrections to the mean-field solution capturing the anisotropy of the free energy with respect to the order parameter orientation.

IV. EVALUATION OF THE FREE ENERGY IN THE GAUSSIAN APPROXIMATION

The first systematic free energy correction is that from *Gaussian fluctuations* about the mean-field solution obtained by expanding the free energy, or equivalently the action, to lowest order in the fluctuation field $\delta\varphi_{\mathbf{q}, \nu}(\tau) = \varphi_{\mathbf{q}, \nu}(\tau) - \varphi_{\mathbf{q}, \nu}^{\text{MF}}$. Introducing the time Fourier transform

$$\varphi_{\mathbf{q}, \nu}(\tau) = \sum_{\omega_n} \varphi_{\mathbf{q}, \nu, \omega_n} \exp[i\omega_n \tau],$$

where $\omega_n = 2\pi n\beta^{-1}$ are bosonic Matsubara frequencies, we immediately get the following bilinear form of the Gaussian fluctuation part of the action:

$$\delta\mathcal{S}\{\delta\varphi_{\mathbf{q},\nu}\} = \beta^{-1} \sum_{\mathbf{q},\nu,\nu'} \sum_{\omega_n} A_{\mathbf{q},\omega_n;\nu,\nu'} \delta\varphi_{\mathbf{q},-\omega_n,\nu}^{(+)*} \delta\varphi_{\mathbf{q},\omega_n,\nu'}^{(+)}, \quad (35)$$

where

$$\delta\varphi_{\mathbf{q},\omega_n,\nu}^{(+)} = \frac{1}{2} [s(\kappa_{\mathbf{q},\nu}) \delta\varphi_{\mathbf{q},\omega_n,\nu} + s^*(\kappa_{\mathbf{q},\nu}) \delta\varphi_{-\mathbf{q},\omega_n,\nu}^*]. \quad (36)$$

Here the fluctuation matrix elements $A_{\mathbf{q},\omega_n;\nu,\nu'}$ describe the weight of the Gaussian fluctuations of wavevector \mathbf{q} , frequency ω_n and polarization ν .

Here a comment is in order. The fields $\delta\varphi$ do not obey the relation $\delta\varphi_{\mathbf{q},\nu}^*(\tau_1) = \delta\varphi_{-\mathbf{q},\nu}(\tau_1)$, i.e. their spatial Fourier transforms are not real-valued, which leads to the combination of fluctuation amplitudes at momenta \mathbf{q} and $-\mathbf{q}$, weighted by the phase factors $s(\kappa_{\mathbf{q},\nu})$. Only symmetric combination of the fields, gives contribution to the free energy because the antisymmetric combination with $\delta\varphi_{\mathbf{q},\omega_n,\nu}^{(-)} = \frac{1}{2} [s(\kappa_{\mathbf{q},\nu}) \delta\varphi_{\mathbf{q},\omega_n,\nu} - s^*(\kappa_{\mathbf{q},\nu}) \delta\varphi_{-\mathbf{q},\omega_n,\nu}^*]$ drops out.

A. Static fluctuations

We start by considering the contribution of static Gaussian fluctuations to the free energy. The expansion of $\mathcal{S}_{loc}^{\text{stat}}$ in terms of fluctuation amplitudes up to second order is given by

$$\begin{aligned} \delta\mathcal{S}_{loc}^{\text{stat}} &= -\beta^{-1} \delta \left\{ \sum_j \ln \cosh(\beta[(\varphi_{\text{MF}} \hat{\mathbf{m}} + \delta\varphi_{i\mu,0})^2]^{1/2}) \right\} \\ &= -\frac{1}{2} \sum_j \{ \beta_c \delta\varphi_{i\mu,0}^2 + \beta_m (\hat{\mathbf{m}} \cdot \delta\varphi_{i\mu,0})^2 \}, \end{aligned} \quad (37)$$

where $\beta_m = (1 - t^2)\beta - \beta_c$ and $\beta_c = \frac{1}{T_c}$. Here $t = \tanh(\beta\varphi_{\text{MF}})$ denotes the dimensionless measure of magnetization, which is zero at T_c and rises monotonically upon cooling to the saturation magnetization ($t = 1$) at $T = 0$. The components of $\delta\varphi_{i\mu,0}$ may be expressed in terms of the momentum space fluctuation amplitudes $\delta\varphi_{\mathbf{q},0,\nu} = \varphi_{\mathbf{q},0,\nu} - \varphi_{\mathbf{q},\nu}^{\text{MF}}$ as

$$\delta\varphi_{i\mu,0}^\alpha = \sum_{\mathbf{q},\nu} \text{Re}\{s(\kappa_{\mathbf{q},\nu}) \chi_{\mathbf{q},\nu;j\alpha}^* \delta\varphi_{\mathbf{q},\nu,0}\}. \quad (38)$$

Now we can rewrite $\delta\mathcal{S}_{loc}^{\text{stat}}$ in the same form as in Eq.(35):

$$\delta\mathcal{S}^{\text{stat}} = \beta^{-1} \sum_{\mathbf{q},\nu,\nu'} A_{\mathbf{q},\nu\nu'}^{\text{stat}} \delta\varphi_{\mathbf{q},0,\nu}^{(+)*} \delta\varphi_{\mathbf{q},0,\nu'}^{(+)}, \quad (39)$$

and the matrix $A_{\mathbf{q},\nu\nu'}^{\text{stat}}$ is found to be

$$A_{\mathbf{q},\nu\nu'}^{\text{stat}} = \beta[(|\kappa_{\mathbf{q},\nu}|^{-1} - \frac{\beta_c}{2})\delta_{\nu\nu'} - \frac{\beta_m}{2} m_{\mathbf{q}\nu} m_{\mathbf{q}\nu'}]. \quad (40)$$

Now the integration over the fluctuation amplitudes may be performed, with the result

$$\begin{aligned} Z^{\text{stat}} &= \frac{Z^{\text{MF}}}{C'} \int [d\varphi] \exp[-\beta\delta\mathcal{S}^{\text{stat}}] \\ &= Z_{\text{MF}} \exp[-\beta\delta\mathcal{F}^{\text{stat}}], \end{aligned} \quad (41)$$

which gives the free energy contribution to be equal to

$$\delta\mathcal{F}^{\text{stat}} = \frac{1}{2} \beta^{-1} \sum_{\mathbf{q}} \ln \det\{A_{\mathbf{q},\nu\nu'}^{\text{stat}}\}. \quad (42)$$

B. Dynamic fluctuations

We now turn to the dynamic fluctuations, obtained by expanding $\mathcal{S}_{loc}^{\text{dyn}}$ to quadratic order in the finite frequency Fourier components $\delta\varphi_{j,\omega_n}$ of the time-dependent fluctuation fields. First we note that $\mathbf{\Omega}_0 = \int d\tau_0 \mathbf{\Omega}(\tau_0)$ may be expressed in terms of the transverse fluctuation amplitudes $\delta\varphi_j^{\text{tr},\alpha}(\tau) = \sum_{\alpha'} P_{\alpha\alpha'} \delta\varphi_j^{\alpha'}(\tau)$, where $P_{\alpha\alpha'} = \delta_{\alpha\alpha'} - m_\alpha m_{\alpha'}$, as

$$\begin{aligned} \mathbf{\Omega}_{j,0} &= \int_0^\beta d\tau \delta\mathbf{e}_j(\tau) \times \delta\dot{\mathbf{e}}_j(\tau) \\ &= \frac{1}{\varphi_{\text{MF}}^2} \int_0^\beta d\tau \delta\varphi_j^{\text{tr}}(\tau) \times \frac{\partial}{\partial\tau} \delta\varphi_j^{\text{tr}}(\tau) \\ &= \frac{\beta_c^2 \beta}{t^2} \sum_{\omega_n} i\omega_n \delta\varphi_{j,-\omega_n}^{\text{tr}} \times \delta\varphi_{j,\omega_n}^{\text{tr}}. \end{aligned} \quad (43)$$

The contribution of the $\mathbf{\Omega}_0$ -term to the action is then given by (taking $\mathbf{e}_j(0) = \hat{\mathbf{m}}$)

$$\delta\mathcal{S}_{loc}^{\text{dyn}} = -\frac{i}{\beta} \sum_j \tanh(\beta\varphi_{\text{MF}}) [\mathbf{\Omega}_{j,0} \cdot \mathbf{e}_j(0)]. \quad (44)$$

It is instructive to write components of the fluctuation amplitudes in the following form ($j = (i, \mu)$)

$$\begin{aligned} \delta\varphi_{i\mu}^\alpha(\tau) &= \sum_{\mathbf{q},\nu=1}^{3N_s} \text{Re}\{s(\kappa_{\mathbf{q},\nu}) N^{-1/2} e^{-i\mathbf{q}\mathbf{R}_i} u_{\mathbf{q},\nu}^{\mu\alpha} \delta\varphi_{\mathbf{q},\nu}(\tau)\} \\ &= \frac{1}{2} N^{-1/2} \sum_{\mathbf{q},\nu=1}^{3N_s} u_{\mathbf{q},\nu}^{\mu\alpha} \{s(\kappa_{\mathbf{q},\nu}) e^{-i\mathbf{q}\mathbf{R}_i} \delta\varphi_{\mathbf{q},\nu}(\tau) \\ &\quad + s^*(\kappa_{\mathbf{q},\nu}) e^{i\mathbf{q}\mathbf{R}_i} \delta\varphi_{\mathbf{q},\nu}^*(\tau)\}, \end{aligned} \quad (45)$$

where we assume real-valued, inversion symmetric eigenfunctions $u_{\mathbf{q},\nu}^{\mu\alpha} = (u_{\mathbf{q},\nu}^{\mu\alpha})^* = u_{-\mathbf{q},\nu}^{\mu\alpha}$. Performing the Fourier transform in time, we get

$$\begin{aligned} \delta\varphi_{i\mu,\omega_n}^\alpha &= \frac{1}{2} \frac{1}{\sqrt{N}} \sum_{\mathbf{q},\omega_n,\nu} u_{\mathbf{q},\nu}^{\mu\alpha} \{s(\kappa_{\mathbf{q},\nu}) e^{-i\mathbf{q}\mathbf{R}_i} \delta\varphi_{\mathbf{q},\omega_n,\nu} \\ &\quad + s^*(\kappa_{\mathbf{q},\nu}) e^{i\mathbf{q}\mathbf{R}_i} \delta\varphi_{\mathbf{q},-\omega_n,\nu}^*\}. \end{aligned} \quad (46)$$

The contribution of the $\mathbf{\Omega}_0$ -term to the local action is then given by

$$\begin{aligned}
\delta S_{loc}^{dyn} &= -\frac{i}{\beta} \frac{(i\beta\beta_c^2)}{t} \sum_{\mathbf{q}, \omega_n, \nu, \nu'} \omega_n D_{\mathbf{q}\nu\nu'} \left[s(\kappa_{\mathbf{q}, \nu}) \delta\varphi_{\mathbf{q}, \omega_n, \nu} s(\kappa_{-\mathbf{q}, \nu'}) \delta\varphi_{-\mathbf{q}, \omega_n, \nu'} + \right. \\
&\quad \left. s^*(\kappa_{\mathbf{q}, \nu}) \delta\varphi_{\mathbf{q}, -\omega_n, \nu}^* s^*(\kappa_{-\mathbf{q}, \nu'}) \delta\varphi_{-\mathbf{q}, -\omega_n, \nu'}^* + s(\kappa_{\mathbf{q}, \nu}) \delta\varphi_{\mathbf{q}, \omega_n, \nu} s^*(\kappa_{\mathbf{q}, \nu'}) \delta\varphi_{\mathbf{q}, -\omega_n, \nu'}^* + s^*(\kappa_{\mathbf{q}, \nu}) \delta\varphi_{\mathbf{q}, -\omega_n, \nu}^* s(\kappa_{\mathbf{q}, \nu'}) \delta\varphi_{\mathbf{q}, \omega_n, \nu'} \right] \\
&= \frac{\beta_c^2}{t} \sum_{\mathbf{q}, \omega_n, \nu, \nu'} \omega_n D_{\mathbf{q}\nu\nu'} \delta\varphi_{\mathbf{q}, -\omega_n, \nu}^{(+)} \delta\varphi_{\mathbf{q}, \omega_n, \nu'}^{(+)*}, \\
D_{\mathbf{q}\nu\nu'} &= \sum_{\mu} \sum_{\alpha_1, \alpha_2, \alpha_3} \sum_{\alpha'_2, \alpha'_3} m_{\alpha_1} \epsilon_{\alpha_1 \alpha_2 \alpha_3} P_{\alpha_2 \alpha'_2} P_{\alpha_3 \alpha'_3} u_{\mathbf{q}\nu}^{\mu \alpha'_2} u_{\mathbf{q}\nu'}^{\mu \alpha'_3}.
\end{aligned} \tag{47}$$

The dynamic fluctuation expression for the exchange interaction term is given by

$$\begin{aligned}
\delta S_{\kappa} &= \frac{1}{\beta} \int_0^\beta d\tau \sum_{\mathbf{q}, \nu} |\kappa_{\mathbf{q}, \nu}|^{-1} \delta\varphi_{\mathbf{q}, \nu}(\tau) \delta\varphi_{\mathbf{q}, \nu}^*(\tau) = \\
&= \sum_{\mathbf{q}, \omega_n \neq 0, \nu} |\kappa_{\mathbf{q}, \nu}|^{-1} \delta\varphi_{\mathbf{q}, \omega_n, \nu}^{(+)} \delta\varphi_{\mathbf{q}, -\omega_n, \nu}^{(+)*}.
\end{aligned} \tag{48}$$

Adding the two contributions we find

$$\delta S^{dyn} = \beta^{-1} \sum_{\mathbf{q}, \omega_n \neq 0; \nu, \nu'} A_{\mathbf{q}, \omega_n; \nu\nu'}^{dyn} \delta\varphi_{\mathbf{q}, 0, \nu}^{(+)*} \delta\varphi_{\mathbf{q}, 0, \nu'}^{(+)}, \tag{49}$$

where $A_{\mathbf{q}, \omega_n; \nu\nu'}^{dyn}$ is given by

$$A_{\mathbf{q}, \omega_n; \nu\nu'}^{dyn} = \beta [|\kappa_{\mathbf{q}, \nu}|^{-1} \delta_{\nu\nu'} + \beta_c^2 t^{-1} \omega_n D_{\mathbf{q}\nu\nu'}]. \tag{50}$$

Now one may perform the integration over the fluctuation amplitudes resulting in

$$Z^{dyn} = Z^{MF} \exp [-\beta \delta \mathcal{F}^{dyn}],$$

which gives the free energy contribution to be equal to

$$\delta \mathcal{F}^{dyn} = \frac{1}{2\beta} \sum_{\mathbf{q}, \omega_n \neq 0} \ln \det \{A_{\mathbf{q}, \omega_n; \nu\nu'}^{dyn}\}. \tag{51}$$

In summary, in the approximation in which the corrections to the free energy come predominantly from Gaussian fluctuations, the partition function is found to be

$$Z = \frac{Z^{MF}}{C'} \exp [-\beta (\delta \mathcal{F}^{stat} + \delta \mathcal{F}^{dyn})].$$

As a sanity check, in the appendix we compute the contribution of dynamic fluctuations to the free energy at low temperature $T \ll T_c$ and show that the contribution of transverse fluctuations from the functional integral representation recovers the spin wave theory result.

V. LIFTING MEAN-FIELD DEGENERACIES IN QUANTUM HEISENBERG-COMPASS SPIN MODEL

Here, we compute the contribution of Gaussian fluctuations to the free energy in the quantum Heisenberg-compass model on the cubic lattice. This model is one of

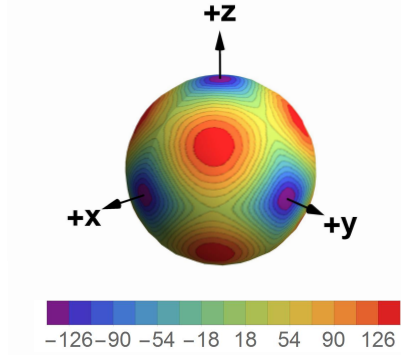


FIG. 1: (Colors online) (a) The magnitude of the Gaussian contribution to the free energy $\delta \mathcal{F}(\theta, \phi)$ computed for $J = -1$, $K = -1.1$ and $\beta = \beta_c + 1$ is plotted on the surface of the unit sphere. The minima of the free energy are shown by deep blue color. The preferred directions of the magnetization are along the cubic axes. The energy scale is shown in units of J .

the simplest models described by Eq. (1), in which the interaction matrix has only diagonal elements:

$$J_{j,j'}^{\alpha\alpha} = \frac{1}{2} \delta_{j', j+\tau_\mu} [J + K \delta_{\alpha, \mu}],$$

where $\tau_\mu = \pm x, \pm y, \pm z$ labels nearest neighbor bonds. The eigenvalues of the exchange operator are given by

$$\kappa_{\mathbf{q}, \nu} = \sum_{\alpha} (J + K \delta_{\alpha, \nu}) \cos q_{\alpha}.$$

The three eigenvectors $\mathbf{u}_{\mathbf{q}\nu}$ point along the three cubic axes, such that the components are $u_{\mathbf{q}\nu}^{\alpha} = \delta_{\nu, \alpha}$, $\alpha = x, y, z$. Provided $J < 0$ and $K < |J|$ the ferromagnetic mean field solution φ_{MF} is given by the solution of the non-linear equation $2|\kappa_0|^{-1} \varphi_{MF} = \tanh(\beta \varphi_{MF})$.

The fluctuation contribution is described by a 3×3 -matrix

$$A_{\mathbf{q}, \omega_n, \nu\nu'} = \delta_{\omega_n, 0} A_{\mathbf{q}, \nu\nu'}^{stat} + (1 - \delta_{\omega_n, 0}) A_{\mathbf{q}, \omega_n, \nu\nu'}^{dyn}, \tag{52}$$

whose matrix elements can be easily obtained for arbitrary orientation of magnetization,

$\hat{\mathbf{m}} = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)$. Using Eq.(40), we get the following expression for the static contribuion:

$$A_{\mathbf{q},\nu\nu'}^{stat} = \begin{pmatrix} d_{\mathbf{q},x} & g_{x,y} & g_{x,z} \\ g_{y,x} & d_{\mathbf{q},y} & g_{y,z} \\ g_{z,x} & g_{z,y} & d_{\mathbf{q},z} \end{pmatrix}, \quad (53)$$

where $d_{\mathbf{q},\nu} = |\beta \kappa_{\mathbf{q},\nu}^{-1}| - b_m m_\nu^2 - b_{tr}$, $g_{\nu\nu'} = -b_m m_\nu m_{\nu'}$, $\kappa_{\mathbf{q},\nu}^{-1} = 1/(3J\gamma_{\mathbf{q}} + K \cos q_\nu)$, $\gamma_{\mathbf{q}} = \frac{1}{3} \sum_\beta \cos q_\beta$, $b_m = \frac{1}{2}(1 - t^2)$, $b_{tr} = \frac{1}{2}\beta_c \beta$. We remind that $t = \tanh(\beta \varphi_{MF})$ is a dimensionless measure of magnetization.

The dynamical matrix is defined by Eq.50, which for the cubic geometry simplifies to the following expression:

$$A_{\mathbf{q},\omega_n,\nu\nu'}^{dyn} = \beta^{-1} [|\kappa_{\mathbf{q},\nu}|^{-1} \delta_{\nu,\nu'} + \omega_n \beta_c^2 t^{-1} \sum_{\alpha_1, \alpha_2, \alpha_3} m_{\alpha_1} \epsilon_{\alpha_1 \alpha_2 \alpha_3} P_{\alpha_2 \nu} P_{\alpha_3 \nu'}] \quad (54)$$

The matrix $A_{\mathbf{q},\omega_n,\nu\nu'}$ may be diagonalized for fixed \mathbf{q}, ω_n . Its eigenvalues $\lambda_{\nu,\mathbf{q},\omega_n} = \lambda_{\nu,\mathbf{q},\omega_n}(\theta, \phi)$ have a rather complex dependence on angles θ and ϕ , implying an angular dependent profile of the fluctuation free energy $\delta\mathcal{F} = \delta\mathcal{F}(\theta, \phi)$. After integration over fluctuations we obtain

$$Z = \frac{Z^{MF}}{C'} \exp[-\beta \delta\mathcal{F}], \quad (55)$$

$$\delta\mathcal{F} = \frac{1}{2\beta} \sum_{\mathbf{q}, \omega_n, \nu} \ln \lambda_{\nu,\mathbf{q},\omega_n} + const.$$

In performing the summation over the Matsubara frequencies, we need to regularize the expression by subtracting a term $\ln[\omega_n \beta_c^2 / \beta t]$ from $\ln \lambda_{\nu,\mathbf{q},\omega_n}$, which will guarantee convergence of the ω_n summation. The subtracted term corresponds to the fluctuation free energy at the transition point.

In Fig.1, we show the angular dependence of $\delta\mathcal{F}(\theta, \phi)$ computed for representative parameters $J = -1$ and $K = -1.1$. The magnitude of $\delta\mathcal{F}(\theta, \phi)$ is presented as a color-coded plot on the unit sphere, where the minima and maxima of the free energy are shown by deep blue and red color, correspondingly. We see that the minima of $\delta\mathcal{F}(\theta, \phi)$ are achieved when the magnetization is directed along one of the cubic axes. This finding clearly shows that while mean field free energy is isotropic, the fluctuation free energy depends upon the direction of the order parameter, indicating that the contribution of fluctuations to the free energy removes the degeneracy of the equilibrium state found on the mean field level.

VI. CONCLUSION

In summary, in this paper we elaborate on a method for calculating the free energy of quantum spin systems using functional integral techniques. We employ a powerful formal technique known as the Hubbard-Stratonovich

transformation to map an interacting quantum spin system into a collection of "single spin"-systems coupled to a fictitious fluctuating magnetic field. This method is very general and can be applied to any biquadratic quantum spin model. Indeed, the Hubbard-Stratonovich transformation applied to isotropic Heisenberg systems in the low temperature limit has been considered before,³⁴⁻³⁶ but has not been applied, as far as we know, to calculate the free energy of anisotropic quantum spin systems. In this work, we present a microscopic derivation of the path-integral representation of the quantum-spin-system partition function for a particular class of quantum spin models with anisotropic bond-directional spin interactions. We determine the contribution of Gaussian fluctuations to the free energy at all temperatures in the ordered phase. Our analysis shows explicitly that the fluctuation free energy has a complex angular dependence, thus breaking the rotational degeneracy of the mean-field ground state.

We believe that the proposed method holds good promise to understand directional ordering in systems with anisotropic bilinear interactions, which are common in SOC systems. In these systems, the high degeneracy of the mean-field solution is lifted by the anisotropy of the spin-spin interaction, such that the spontaneous magnetization is pinned along certain preferred directions. The latter may change with temperature.

For illustration, we apply the above analysis to the quantum Heisenberg-compass spin model and show that the direction of the order parameter in spin space is selected by fluctuations and is determined by the competition between Heisenberg and compass terms. For the range of parameters for which the ferromagnetic state is the ground state, the Gaussian fluctuations select the cubic axes as directions of the magnetization.

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Appendix A: Isotropic Heisenberg model

Here we calculate the contribution of dynamic fluctuations to the free energy at low temperature $T \ll T_c$. It

is known that the leading contribution comes from spin wave excitations. The purpose of this appendix is to show that the contribution of transverse fluctuations from the functional integral representation recovers the spin wave theory result.

As a simple example we calculate the contribution of dynamic fluctuations for the case $\kappa_{\mathbf{q},\nu} = 3J\gamma_q$, where $\gamma_q = \frac{1}{3} \sum_{\alpha} \cos q_{\alpha}$ and $J < 0$. The spin wave excitation energy in our representation is given by

$$\omega_{\mathbf{q}} = \frac{t}{\beta_c^2 |\kappa_{\mathbf{q}=0}| |\kappa_{\mathbf{q},\nu}|} \left[|\kappa_{\mathbf{q}=0}| - |\kappa_{\mathbf{q},\nu}| s^2(\kappa_{\mathbf{q},\nu}) \right]. \quad (\text{A1})$$

In the limit $q \ll 1$, it can also be significantly simplified:

$$\omega_{\mathbf{q}} \approx \frac{3}{4} |J| [1 - \gamma_q]. \quad (\text{A2})$$

In this limit, the dynamic fluctuation matrix then takes the form

$$A_{\mathbf{q},\omega_n;\nu,\nu'}^{\text{dyn}} = \frac{\beta_c^2}{\beta t} \begin{pmatrix} \omega_{\mathbf{q}} & \omega_n \\ -\omega_n & \omega_{\mathbf{q}} \end{pmatrix}. \quad (\text{A3})$$

Its determinant is equal to

$$\det\{A_{\mathbf{q},\omega_n;\nu,\nu'}^{\text{dyn}}\} = \left(\frac{\beta_c^2}{\beta t}\right)^2 [\omega_n^2 + \omega_{\mathbf{q}}^2]. \quad (\text{A4})$$

Recalling that the transverse fluctuation free energy is given by

$$\delta\mathcal{F}^{\text{tr}} = \frac{1}{2\beta} \sum_{\mathbf{q},\omega_n \neq 0} \ln \det\{A_{\mathbf{q},\omega_n;\nu,\nu'}^{\text{dyn}}\},$$

the contribution to the partition function is found to be

$$\begin{aligned} Z^{\text{tr}} &= \exp[-\beta\delta\mathcal{F}^{\text{tr}}] = \exp\left[-\frac{1}{2} \sum_{\mathbf{q},\omega_n \neq 0} \ln \det\{A_{\mathbf{q},\omega_n;\nu,\nu'}^{\text{dyn}}\}\right] \\ &= \exp\left[-\frac{1}{2} \sum_{\mathbf{q},\omega_n \neq 0} \{\ln(\beta^{-1}\beta_c^2 t^{-1})^2 + \ln[\omega_n^2 + \omega_{\mathbf{q}}^2]\}\right]. \end{aligned} \quad (\text{A5})$$

The first term in the curly brackets give simple constant renormalization. The summation over Matsubara frequencies in the second term gives

$$X_{\mathbf{q}} = \text{Re} \frac{1}{2} \sum_{\omega_n} \ln[\omega_n^2 + \omega_{\mathbf{q}}^2] = \text{Re} \frac{1}{2} \sum_{\omega_n} \ln[(i\omega_n)^2 - \omega_{\mathbf{q}}^2] \quad (\text{A6})$$

$$= \frac{1}{2} \beta \omega_{\mathbf{q}} + \ln[1 - \exp(-\beta \omega_{\mathbf{q}})], \quad (\text{A7})$$

which leads to the free energy contribution

$$\delta\mathcal{F}^{\text{tr}} = \frac{1}{2} \sum_{\mathbf{q}} \{\omega_{\mathbf{q}} + 2\beta^{-1} \ln[1 - \exp(-\beta \omega_{\mathbf{q}})]\} + \text{const.} \quad (\text{A8})$$

To get this result we differentiate $X_{\mathbf{q}}$ with respect to $\omega_{\mathbf{q}}$

$$\frac{\partial}{\partial \omega_{\mathbf{q}}} X_{\mathbf{q}} = \frac{1}{2} \sum_{\omega_n} \left[\frac{1}{i\omega_n + \omega_{\mathbf{q}}} - \frac{1}{i\omega_n - \omega_{\mathbf{q}}} \right] \quad (\text{A9})$$

$$= \frac{1}{2} \beta [-n_B(-\omega_{\mathbf{q}}) + n_B(\omega_{\mathbf{q}})] \quad (\text{A10})$$

where $n_B(\omega_{\mathbf{q}}) = [\exp(\beta \omega_{\mathbf{q}}) - 1]^{-1}$ is the Bose distribution function.

The term $\sum_{\mathbf{q}} \omega_{\mathbf{q}}$ is the zero point fluctuation contribution of the two transverse modes to the ground state energy (note that $\omega_{\mathbf{q}}$ is independent of ν for the isotropic model considered). There must be an additional constant contribution $\delta\omega_0$ to the ground state energy, which is not completely captured by the Gaussian fluctuation contribution, such that $\sum_{\mathbf{q}} \omega_{\mathbf{q}} + \delta\omega_0 \propto \sum_{\mathbf{q}} \gamma_{\mathbf{q}}$, which sums to zero. Recall that for the isotropic model the ground state is identical to the mean field ground state, such that the fluctuation contribution to the ground state energy vanishes. The fluctuation contribution to the internal energy is then given

$$\delta U^{\text{dyn}} = \delta\mathcal{F}^{\text{dyn}} + \beta \frac{\partial \delta\mathcal{F}^{\text{dyn}}}{\partial \beta} = \sum_{\mathbf{q}} \omega_{\mathbf{q}} \left\{ \frac{1}{2} + n_B(\omega_{\mathbf{q}}) \right\} \quad (\text{A11})$$

This is identical with the standard result of spin wave theory, except that $\omega_{\mathbf{q}}$ differs from the spin wave result at higher q . At low temperatures δF^{dyn} provides the leading contribution to the thermodynamic quantities, e.g. $\delta U^{\text{dyn}} \propto T^{5/2}$, whereas the longitudinal fluctuations contribute an exponentially small term. As $\omega_{\mathbf{q}} = \frac{1}{2} |J| q^2 + O(q^4)$, the leading low temperature behavior of δU^{dyn} agrees exactly with the conventional spin wave result.

¹ G. Khaliullin, Prog. Theor. Phys. Suppl. **160**,155 (2005).

² G. Jackeli and G. Khaliullin, Phys. Rev. Lett. **102**, 017205 (2009).

³ J. Chaloupka, G. Jackeli, and G. Khaliullin, Phys. Rev. Lett. **105**, 027204 (2010).

⁴ N. B. Perkins, Y. Sizyuk and P. Wölfle, Phys. Rev. B **89**,

- 035143 (2014).
- ⁵ Y. Sizyuk, C. Price, P. Wölfle, and N. B. Perkins, *Phys. Rev. B* **90**, 155126 (2014).
 - ⁶ J. G. Rau, E. Kin-Ho Lee, H.-Y. Kee, *Phys. Rev. Lett.* **112**, 077204 (2014).
 - ⁷ I. Kimchi and A. Vishwanath, *Phys. Rev. B* **89**, 014414 (2014).
 - ⁸ Z. Nussinov, J. van den Brink, *Rev. Mod. Phys.* **87** 1 (2015).
 - ⁹ H. Ishizuka and L. Balents, *Phys. Rev. B* **90**, 184422 (2014).
 - ¹⁰ J. Chaloupka and G. Khaliullin, *Phys. Rev. B* **92**, 024413 (2015).
 - ¹¹ J. Chaloupka and G. Khaliullin, *Phys. Rev. B* **94**, 064435 (2016).
 - ¹² I. Rousochatzakis, J. Reuther, R. Thomale, S. Rachel and N. B. Perkins, *PRX* **5**, 041035 (2015).
 - ¹³ I. Rousochatzakis and N. B. Perkins, arXiv:1610.08463.
 - ¹⁴ Michael Becker, Maria Hermanns, Bela Bauer, Markus Garst, Simon Trebst, *Phys. Rev. B* **91**, 155135 (2015).
 - ¹⁵ G. Jackeli and A. Avella, *Phys. Rev. B* **92**, 184416 (2015).
 - ¹⁶ J. G. Rau, E. Kin-Ho Lee, H.-Y. Kee, *Annual Review of Condensed Matter Physics* **7**, 195 (2016).
 - ¹⁷ A. Kitaev, *Ann. Phys.* **321**, 2 (2006).
 - ¹⁸ S. K. Choi, R. Coldea, A. N. Kolmogorov, T. Lancaster, I. I. Mazin, S. J. Blundell, P. G. Radaelli, Yogesh Singh, P. Gegenwart, K. R. Choi, S.-W. Cheong, P. J. Baker, C. Stock, and J. Taylor, *Phys. Rev. Lett.* **108**, 127204 (2012).
 - ¹⁹ S. H. Chun, J.-W. Kim, J. Kim, H. Zheng, C. C. Stoumpos, C. D. Malliakas, J. F. Mitchell, Kavita Mehlawat, Yogesh Singh, Y. Choi, T. Gog, A. Al-Zein, M. Moretti Sala, M. Krisch, J. Chaloupka, G. Jackeli, G. Khaliullin, B. J. Kim, *Nature Physics* **11**, 462 (2015).
 - ²⁰ A. Banerjee, C.A. Bridges, J.-Q. Yan, A.A. Aczel, L. Li, M.B. Stone, G.E. Granroth, M.D. Lumsden, Y. Yiu, J. Knolle, D.L. Kovrizhin, S. Bhattacharjee, R. Moessner, D.A. Tennant, D.G. Mandrus, S.E. Nagler, *Nature materials* **15**, 733 (2016).
 - ²¹ R. D. Johnson, S. Williams, A. A. Haghighirad, J. Singleton, V. Zapf, P. Manuel, I. I. Mazin, Y. Li, H. O. Jeschke, R. Valenti, R. Coldea, *Phys. Rev. B* **92**, 235119 (2015).
 - ²² A. Biffin, R. D. Johnson, S. Choi, F. Freund, S. Manni, A. Bombardi, P. Manuel, P. Gegenwart, and R. Coldea, *Phys. Rev. B* **90**, 205116 (2014).
 - ²³ A. Biffin, R.D. Johnson, I. Kimchi, R. Morris, A. Bombardi, J.G. Analytis, A. Vishwanath, and R. Coldea, *Phys. Rev. Lett.* **113** 197201 (2014).
 - ²⁴ T. Takayama, A. Kato, R. Dinnebier, J. Nuss, H. Kono, L.S.I. Veiga, G. Fabbri, D. Haskel, and H. Takagi, *Phys. Rev. Lett.* **114**, 077202 (2015).
 - ²⁵ Y. Sizyuk, P. Wölfle and N. B. Perkins, *Phys. Rev. B* **94**, 085109 (2016).
 - ²⁶ J. Villain, R. Bidaux, J.-P. Carton, and R. Conte, *J. Phys. France* **41**, 1263 (1980).
 - ²⁷ E. F. Shender, *Sov. Phys. JETP* **56**, 178 (1982).
 - ²⁸ A. Chubukov, *Phys. Rev. Lett.* **69**, 832 (1992).
 - ²⁹ M. E. Zhitomirsky, M. V. Gvozdkova, P. C. W. Holdsworth, and R. Moessner, *Phys. Rev. Lett.* **109**, 077204 (2012).
 - ³⁰ G.-W. Chern and R. Moessner *Phys. Rev. Lett.* **110**, 077201 (2013).
 - ³¹ P.A. McClarty, P. Stasiak, and M. J. P. Gingras, *Phys. Rev. B* **89**, 024425 (2014).
 - ³² A. L. Chernyshev and M. E. Zhitomirsky, *Phys. Rev. Lett.* **113**, 237202 (2014).
 - ³³ A. L. Chernyshev and M. E. Zhitomirsky, *Phys. Rev. B* **92**, 144415 (2015).
 - ³⁴ A. Angelucci and G. Jug, *Int. J. Mod. Phys. B* **3**, 1069 (1989).
 - ³⁵ A. Angelucci, *Phys. Rev. B* **44**, 6849 (1991).
 - ³⁶ A. Angelucci, *Phys. Rev. B* **45**, 5387 (1992).
 - ³⁷ J. W. Negele and H. Orland, *Quantum Many Particle Systems* Westview Press, Boulder (1998).
 - ³⁸ E. Fradkin, *Field Theories of Condensed Matter Physics* Cambridge University Press, Cambridge, UK (2013).
 - ³⁹ J. Hubbard, *Phys. Rev. Lett.* **3**, 77 (1959).
 - ⁴⁰ R. L. Stratonovich, *Sov. Phys. Dokl.* **2**, 416 (1958).
 - ⁴¹ Y. Sizyuk, N. B. Perkins and P. Wölfle, *Phys. Rev. B* **92**, 155131 (2015).
 - ⁴² H. Trotter, *Proc. Am. Math. Soc.* **10**, 545 (1959).